The Influence of Monomolecular Surface Films on the Production of Condensation Nuclei From Bubbled Sea Water

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ABSTRACT

Salt condensation nuclei were produced in large quantities (10^5 parti-
cles per cm^3) from bubbled sea water and sodium chloride solutions. The
addition of pure and mixed insoluble m o n o m o l e c u l a r films to the sea-
water surface increased the concentration of salt nuclei by as much as
threefold. The greatest increases were caused by the binary monolayers,
which were synergistic in their behavior. No increases in nuclei count
resulted when monolayers were spread onto the surface of aqueous sys-
tems which did not contain surface-active foam-forming materials.

The increase in the number of salt particles measured is not related
to a surface-chemical modification of either the sea-water droplets or the
surface of the salt particles but is a consequence of the alteration of the
mechanics of the bubble-bursting process. The insoluble film decreases
the degree of foaming at the sea-water surface and enhances the immedi-
ate breaking of small bubbles. This leads to a greater rate of surface
fragmentation of the air/water interface. The meteorological and oceano-
graphic implications of these data are discussed.

PROBLEM STATUS

This is an interim report; work on this problem is continuing.

AUTHORIZATION

THE INFLUENCE OF MONOMOLECULAR SURFACE FILMS ON THE PRODUCTION OF CONDENSATION NUCLEI FROM BUBBLED SEA WATER

INTRODUCTION

Bursting of air bubbles and the generation of sea spray are important microscale processes at the air/sea interface. The sea-water droplets projected into the marine atmosphere provide a source of salt nuclei which are thought to be instrumental in the initiation of rain from nonfreezing clouds (1). The ejected jet drops from the bursting bubble have also been shown to effect a sea-to-air transport of surface-active organic material (2). After the sea-water droplets have evaporated, the remaining airborne salt particles may be coated with an organic film which could reduce the surface wettability. The cloud-droplet nucleating efficiency would then be decreased, since the particle surface would have a nonzero contact angle to water, and a greater cloud supersaturation would be required for the nucleation of a water droplet (3).

Another possible consequence of airborne surface-active organic material is the reduction of the evaporation rate of film-coated droplets (4, 5). This effect becomes significant when the molecules of the droplet surface film become close-packed on the shrinking droplet surface as evaporation occurs. Although the foregoing literature dealt specifically with a well-known agent for evaporation retardation (cetyl alcohol), Goetz (6) postulates that as a droplet evaporates natural organic films would concentrate at the air/water interface creating a barrier to evaporation. This process, he concludes, would stabilize fogs at lower humidities than normal, creating a rather permanent haze of high organic content.

The ocean may contain both soluble and insoluble surface-active material at the air/sea interface which can be incorporated into the bubble-generated sea-water aerosols. The constitution of the insoluble slick-forming substances (7) and the properties of naturally occurring surface films have been characterized (8). Although the modification of the properties of salt nuclei and cloud droplets by surface films from the sea have been discussed in theory, few definitive experiments have been conducted to demonstrate such processes. Therefore, the present experiments were designed to: (a) generate salt nuclei by bubbling filtered air through natural and treated sea waters, (b) measure the number of nuclei produced, and (c) determine the effect of surface-active additives.

APPARATUS AND METHODS

The major components of the experimental system (Fig. 1) were the bubbler (salt nuclei generator) and the condensation nuclei counter (Gardner Associates, Inc.). Air was pumped through the system at controlled rates by a tubing pump with electronic speed control. Two cotton-packed columns in series served to provide particle-free air to the glass bubble-nuclei generator, which contained a glass gas-diffuser frit. The filtered air gave nuclei counts of 0, even though the room air counts averaged 10^4 counts/cm^3. The experimental counts of the air passing through the filters and bubbler system were due solely to salt nuclei being generated in the bubble chamber.

To determine the influence of flow rate upon the nuclei production by the generator, filtered air was passed through the system at various rates (0.10 to 0.96 l/min). The
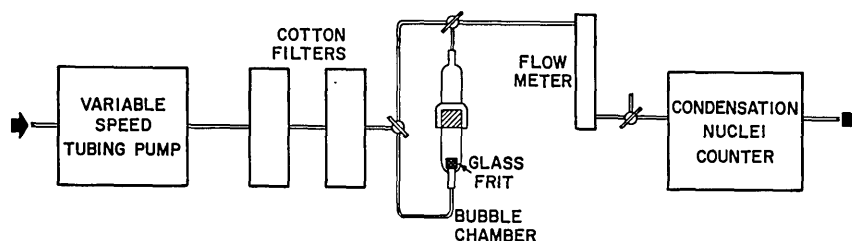
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Fig. 1 — Experimental system for the generation and measurement of condensation nuclei from bubbled sea water

bubbler contained 400 ml of a bucket sample of sea water collected 24 km east of Ocean City, Maryland. A graph of airflow rate vs the count of salt nuclei produced (Fig. 2) shows that the concentrations of nuclei increase markedly at the lower flow rates up to about 0.3 l/min. Above 0.3 l/min, the salt nuclei concentrations increase much more slowly with increasing flow rate. A flow rate of 0.39 l/min was used in most of the experiments reported unless otherwise stated. The upper plot represents the nuclei production from the same sea-water system whose surface contained a monomolecular film of oleic acid. The results in this case are similar to the fresh sea-water experiment, although the nuclei counts are somewhat higher in the presence of the fatty acid monolayer.
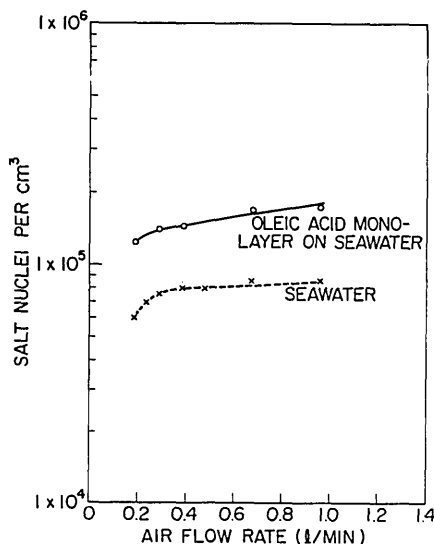

Fig. 2 — Influence of airflow rate on salt nuclei production

     The condensation nuclei counter was equipped with a variable expansion chamber, which theoretically counts particles of various sizes depending upon the supersaturations produced by the expansion of the sampled air which had been saturated with water vapor in the collection chamber. (The nuclei count is measured by the attenuation of light caused by droplets grown from the nuclei in the expansion chamber). However, no particle-count differences were obtained within the variability of relative humidities (101 to 400%) produced by the expansion settings of the instrument. This result was

obtained for both fresh and monolayer-covered sea-water surfaces and may be explained by assuming that all of the counted nuclei were larger than 1.3×10^{-5} cm radius, the largest water-insoluble particle discriminated by the counter. However, the hygroscopic nature of salt particles may make even smaller ones active condensation nuclei at the lower supersaturations. Twomey (9) has derived a relationship between the supersaturation and the minimum radius required for a soluble NaCl particle to be capable of nucleating. From this expression it was calculated that the salt particles counted in this experiment had a radius of greater than 1.16×10^{-6} cm.

The materials used in this study included:

1. sea water collected 24 km east of Ocean City, Maryland (bucket sample),
2. 3% NaCl solution prepared from C.P. grade NaCl and triply distilled water,
3. oleic acid (white), California Corp. for Biochemical Research, freshly distilled at NRL,
4. triolein (glyceryl trioleate), Mann Research Laboratories, Inc., >99%,
5. cetyl alcohol (1-hexadecanol), Lachat Chemical Co., >99%,
6. ethyl palmitate, melting point 21-23°C, Fisher Scientific Co.,
7. cottonseed oil, U.S.P., Fisher Scientific Co., approximate composition 97% mixed triglyceryl esters of fatty acids (primarily palmitic, oleic, and linoleic), iodine value 106-113, and
8. polydimethylsiloxane (dodecamer), molecular weight 978.1 g/mole, Dow Corning.

EFFECT OF MONOMOLECULAR SURFACE FILMS

The number of condensation nuclei counted by the Gardner instrument for fresh untreated sea water is compared with similar data for the same sea water onto which a surface film of oleic acid had been spread (Fig. 2). Following the bubbling and measurement of salt particles from the sea water, a small drop (5 mg) of the monolayer-forming material was placed on the water surface and counts were made under identical bubbling conditions. Only a small portion of the drop spread over the sea water until the surface contained a monomolecular layer of the surface-active substance. The remainder of the drop existed as a liquid lens which did not spread until part of the monolayer was removed. Thus, the insoluble monolayer-forming material does not spread upon its own monomolecular film but provides a source of material to regenerate monolayer removed by the bursting bubbles.

The effect of the monolayer (Fig. 2) is to increase the concentration of salt nuclei detected by the counter. The oleic acid film produced an approximate increase in the nuclei concentration of 50%. Similar results were obtained when this surface-active material was spread on the 3% NaCl solution. All other monolayers caused increases in nuclei production in varying degrees. Data for pure and mixed films are listed in Table 1 for both fresh sea water and the 3% NaCl solution. Both clean distilled water and distilled water covered by an oleic acid film yielded very small nuclei counts at a flow rate of 0.39 l/min. Consequently, there was no measurable contribution from the monolayer itself to the nuclei count. The average deviation of the data in Table 1 ranged from 2 to 4% and was seldom higher than ±5%. Day-to-day average deviation of counts of the untreated aqueous substrates was ±5%. The last column in Table 1 is the ratio of the nuclei count from a monolayer-covered surface to that for the untreated water. In each experiment counts of the fresh water were determined prior to the addition of the surface film.

Similar results were obtained for oleic acid on sea water and on the 3% NaCl solution. This similarity is probably a fortuitous result and indicates that the two substrates possessed similar initial surface activity. Even though C.P. grade chemicals were used in

Table 1

Increase in Nuclei Concentration Due to Monomolecular Films

Monolayer	Substrate	Salt Nuclei Concentration per cm^3*	Ratio of concentration with monolayer to concentration without monolayer
Air through cotton filters only			
None	Distilled water	5×10^2	—
Oleic acid	Distilled water	5×10^2	—
None	Fresh sea water	9×10^4	—
Oleic acid	Fresh sea water	1.4×10^5	1.56
Glyceryl trioleate	Fresh sea water	1.3×10^5	1.44
Mixed film of oleic acid and glyceryl trioleate	Fresh sea water	1.4×10^5	1.56
Cottonseed oil	Fresh sea water	1.2×10^5	1.33
Mixed film of oleic acid and cottonseed oil	Fresh sea water	1.9×10^5	2.11
Cetyl alcohol	Fresh sea water	1.2×10^5	1.33
Mixed film of oleic acid and cetyl alcohol	Fresh sea water	1.9×10^5	2.11
Dodecanol-1	Fresh sea water	1.45×10^5	1.61
Dimethylsiloxane (dodecamer)	Fresh sea water	1.3×10^5	1.44
Mixed film of dodecanol-1 and dimethylsiloxane	Fresh sea water	3.0×10^5	3.33
None	3% NaCl	1.2×10^5	—
Oleic acid	3% NaCl	1.9×10^5	1.58
Cottonseed oil	3% NaCl	1.6×10^5	1.33
Mixed film of oleic acid and cottonseed oil	3% NaCl	2.6×10^5	2.17
Ethyl palmitate	3% NaCl	1.45×10^5	1.21
Mixed film of cottonseed oil and ethyl palmitate	3% NaCl	1.45×10^5	1.21
Cetyl alcohol	3% NaCl	1.75×10^5	1.46
Mixed film of oleic acid and cetyl alcohol	3% NaCl	2.2×10^5	1.83

* Average for five or more measurements

the preparation of the salt solution, surface-active substances which are adsorbed onto the solid salt are extremely difficult to eliminate. Filtration of small volumes through 0.1-μ membrane filters proved a simple technique for the removal of most of the surface-active impurities.

Increases in nuclei production varied from 33 to 61% for single-component monolayers. Premixed films consisting of equal quantities by volume of two components caused greater increases in most cases. A 233% increase resulted when a mixed film of the silicone oil and dodecanol was spread on the sea water. The nuclei counts of most mixed films were higher than counts with either component monolayer by itself. Exceptions to this synergism were the mixed films of cottonseed oil and ethyl palmitate on the NaCl solutions and oleic acid with triolein on sea water. No increase in nuclei count over that for either component was found as a result of mixing these two components.

DISCUSSION

Origin of Condensation Nuclei

The concentrations of nuclei produced by this system are large (approximately 10^5 per cm^3), and it is possible to derive some information about the mechanics of bubble bursting from these data. The bursting of single air bubbles has been studied in detail (10, 11) revealing at least two distinct modes of breakup. The initial rupture of the bubble film or crown produces a number of tiny fragments, which are generally 2 to 10 μ in diameter. These film drops are ejected several millimeters into the air.. The hydrodynamic collapse of the remaining bubble cavity projects several "jet drops" as much as 19 cm above the water surface. These latter drops have diameters in the neighborhood of 100μ for a 1-mm-diameter bubble. Further tearing of the bubble film by the upward rushing jet generates additional film drops. The small film drops may number in the hundreds for bubbles of several millimeters in diameter.

The large number of salt particles produced by the bubbler used in this study cannot be accounted for solely by a jet-drop mechanism. The bubbles generated by the glass frit were approximately 1 mm in diameter. From the number of bubbles which produced 9×10^4 particles per cm^3 using the fresh sea water, it was estimated crudely that about 50 particles resulted from each bubble burst. This estimation is in good agreement with the upper limit of Blanchard's data for the number of film drops created by the bursting of a 1-mm-diameter bubble.

Preining, et al. (12), have determined the concentrations of salt nuclei produced from bubbles of NaCl solutions as a function of concentration. Above 1% NaCl, the salt nuclei concentration was not significantly dependent upon the salinity. Thus, small variations in salinity from one sea-water sample to another would yield negligible differences in nuclei concentrations produced by a particular bubbler system. However, from the data in Table 1, it is apparent that the nuclei concentrations would be greatly influenced by the surface chemical condition of the sea-water sample.

Effect of Surface Films

One of the primary objectives of this research was to test the proposition that organic films might deactivate or poison airborne particles and decrease their efficiency as condensation nuclei (13). If deactivation had been caused by the presence of the surface film adsorbed onto the salt particles, we would have expected to find a decrease in the total number of nuclei from that produced from the untreated sea water. Another consequence of the organic film might be a slowing of the droplet growth rate in the

nuclei counter. This latter mechanism would also lead to an apparent decrease in count, since less light attenuation would occur in the counter tube from the smaller drops. Whether or not the above possibilities should have been expected at the high supersaturations used in the particle counter, the increase in nuclei which was caused by each of the organic films was not immediately anticipated.

Furthermore, these data cannot be explained by surface adsorption of fragments of the monomolecular film onto the hydrated salt particles. It was calculated that there was enough organic material available in these experiments to cover the surfaces of 10^5 particles per cm and form an effective hydrophobic layer. However, film-coated nuclei would not lead to an increase in number or final size of the droplets formed in the nuclei counter.

The most plausible explanation of the data is that the increase in the number of particles produced is a direct result of the presence of an insoluble surface film at the air/water interface. A mechanism to explain the increase in particle concentration is related to the bubble stability characteristics imparted by surface films (14). At high film pressures, most insoluble monolayers formed rigid films above the bubble, reduced bubble lifetimes even in the presence of foam-forming substances, and acted as a foam-breaking agent. In the present study it was observed that there was a 1-cm head of foam at the top of the bubbled sea water. The extent of foaminess agreed in a qualitative and inverse manner with the number of nuclei measured. Breaking of the foam coincided with increased salt-particle counts.

When oleic acid, dodecanol, and the synergistic binary monolayers were present, there was no stabilized foam at the sea-water surface. Thus, in the presence of these monolayers, air bubbles burst almost instantaneously upon reaching the surface. By contrast, the fresh sea-water surface was foamy; many bubbles aggregated and formed a few large bubbles before the surface ruptured. The total surface area involved in breaking per unit time (number of bubbles bursting) was greater in the presence of the insoluble monolayer. The number of surface fragments which eventually become salt nuclei was increased as a result of the altered mechanical properties of surface imparted by the monomolecular films.

An air bubble acts as a transport agent for surface-active material, collecting it from the sea by adsorption onto the air bubble/water interface as it rises. The material may then be deposited at the sea surface or ejected into the marine atmosphere when the bubble bursts. An experiment was performed to examine the change in nuclei production with time as an indication of chemical changes taking place at the air/sea interface as a result of these bubble transport processes (Fig. 3). A smaller bubble generator (62 ml sea water) was constructed which could be easily disassembled for the addition or removal of surface films. During the first phase of the experiment, bubbling (0.2 l/min) of the fresh sea water transported natural insoluble surface-active material to the surface, where it gradually replaced the more soluble foam-generating substances. Nuclei counts increased slightly and foaminess at the sea-water surface decreased. The preferential adsorption of insoluble organic film-forming materials and the exclusion of the more soluble species occurs in sea-surface films (8). After 45 min of bubbling, the dynamic chemical-exchange properties at the sea-water surface were in equilibrium with the arriving and bursting bubbles, and the condensation nuclei count became constant.

A small quantity of oleic acid (0.083 mg) was then added to the sea-water surface using a dilute chloroform spreading solution. In this way a complete monolayer was formed, but a large excess of the surface-active agent was avoided. Under these conditions, a few minutes were required for the insoluble oleic acid to incorporate into and
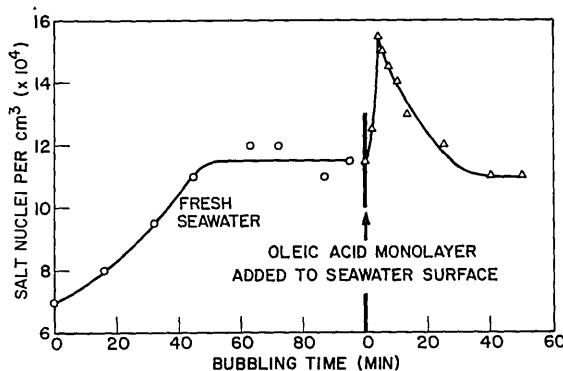
Fig. 3 — Changes in salt nuclei concentra-
tion from bubbled sea water (-o-) and the
effect of adding a small quantity of oleic
acid (83 μg) (-$\triangle$-)

displace other material from the surface. Once displacement had occurred, the nuclei count had risen to 1.55×10^5, which represented an increase of 35% above the count at the time of monolayer addition and 121% greater than the number of nuclei produced from the fresh sea water.

Additional bubbling caused the nuclei count to decrease from its maximum of 1.55×10^5 to the value measured prior to the addition of the oleic acid. Monolayer molecules were being removed by the fragments of the bursting bubbles, which were projected into the atmosphere. It was also considered possible that the disturbance at the sea-water surface caused by the bubbling might be submerging and dissolving the monolayer. A period of 30 min without bubbling was allowed at the conclusion of the previously described experiment. When bubbling was recommenced, a nuclei count of 1.2×10^5 was obtained. Since this value agreed closely with the final value (Fig. 3), it appears that most of the surface film had actually been lost to the air above the bubbler rather than submerged by agitation into the water.

It should be emphasized that the data presented in this report are not necessarily reproducible from one sea-water sample to another. The increases in salt nuclei concentration are primarily dependent upon the nature and quantity of surface-active organic material in the water. Both soluble foam-stabilizing and foam-breaking material are usually present in varying relative amounts. In this regard several experiments were performed to demonstrate the effect of filtration upon the chemical effects at the sea-water surface which affect the nuclei production rate. Fresh sea water was filtered through 0.1-μ Millipore (type VC) filters prior to bubbling. Nuclei concentrations from the filtered water were 1.35×10^5 per cm^3 compared with 7×10^4 per cm^3 for the fresh sea water. This result is equivalent to the combined effects (Fig. 3) of bubbling the fresh sea water and the addition of an insoluble monolayer. When a small drop of oleic acid was added to the filtered water, the nuclei count remained unaltered. It is likely that filtration removed most of the foam-stabilizing substances. The filter had removed not only particulate substances but also soluble surface-active material by adsorption onto the filter fibers. Thus, little foaminess occurred in the filtered water, and the addition of an insoluble monolayer caused no decrease in foam or increase in the nuclei-production rate. This is further proof that the additive surface film does not chemically alter the surface of the salt nuclei but affects the nuclei concentration produced by modifying the bubble-breaking properties.

A similar experiment was performed on the 3% NaCl solution using the smaller bubbling chamber and a flow rate of 0.20 l/min. When the clear salt solution had been filtered twice through $0.1-\mu$ filters, it behaved as if it were essentially free of surface-active material. There was no foaming tendency, bubbles burst instantaneously upon reaching the water surface, and a nuclei count of 1.45×10^5 per cm^3 was measured. The addition of an oleic acid monolayer did not alter the count. By contrast, the unfiltered salt solution yielded a nuclei count of 8×10^4 per cm^3 and was characterized by a 0.5-cm head of foam. The added oleic acid monolayer eliminated the foam and increased the nuclei count to 1.5×10^5 per cm^3, a value about that of the filtered solution. As in previous studies, the results were not dependent upon the supersaturations (1 to 300%) in the cloud chamber.

SEQUEL

Blanchard (10) has studied single bubbles bursting at a sea-water surface covered by an oleic acid film. No film drops were observed in a thermal diffusion cloud chamber into which the fragments of the burst bubble were projected. However, if several bubbles were allowed to cluster before breaking, the number of film drops produced was not diminished by the presence of the monolayer. Although no explanation for these observations was offered, it was concluded that the numerous bubble clusters formed immediately after the breaking of a wave would be independent of surface film properties. On the other hand, the nuclei production from single bubbles which subsequently reach the surface would be affected by the presence or absence of an insoluble monolayer at the sea surface.

The present research deals primarily with multiple bubble bursting (clusters) at a rate which can generate surface foams. Few individual bursts occurred in this system due to the large number reaching the water surface at any instant. Thus, it would not be expected that in this experiment the surface film would cause the film drop decreases observed by Blanchard (10).

At sea, when large quantities of air had been temporarily entrapped by breaking waves or precipitation, many air bubbles would reach the surface simultaneously. In this case the presence of a pre-existing insoluble monolayer (sea slick) would reduce bubble stability (14) and produce more bubble fragments (film and jet drops). This effect would be especially pronounced in sea water that is rich in (foam-stabilizing) surface-active material.

The worldwide influence of natural sea slicks (generally composed of insoluble surface-active material (7)) on the concentration of atmospheric salt nuclei is probably quite small. Natural surface films occur primarily in the biologically active coastal waters and are rapidly dispersed, collapsed, or solubilizied by wind and wave action of sufficient intensity to cause white caps and froth from breaking waves. Therefore, oceanographic and meteorological conditions which produce intensive bubble entrapment would dissipate a monomolecular surface film (sea slick). Furthermore, since the molecules responsible for a sea slick are not usually compressed to their fullest extent (15) (greatest film pressure), their foam-breaking property would not be at a maximum (14). As a consequence of this, there would be a smaller influence of a natural sea slick at lower film pressures upon the production of salt nuclei from bubble bursting.

These data further suggest that surface-active material transported into the marine atmosphere by the ejected film and jet drops does not significantly alter the surface physics of the resulting salt particles. There was no apparent reduction in the nucleating efficiency of the particles at supersaturations from 1 to 300%. While this

proposition has not been conclusively demonstrated, the finding is in accord with the results of Baer, et al. (16). These authors found that although the wettability of certain metal surfaces was drastically altered by adsorbed surface-active materials (determined by liquid-water-drop contact angles), active sites which promote nucleation of vapor condensation were not totally covered by immersion in surfactant solutions. Thus, the solid surface appeared to be coated, but in reality, uncovered areas existed which were of sufficient size to accommodate water-vapor molecules and act as sites for initiation of vapor condensation. If these results can be applied to the case of the salt nuclei surface, the likelihood of total molecular coverage by airborne organic material is small, and nucleation inhibition by vapor blocking would be a difficult condition to achieve with adsorbed surface films.

The deactivation of a potential condensation or freezing nucleus is an important consideration for the cloud physicist in his quest for useful weather modifications. Whether this may be achieved on a sufficiently large scale through the vapor-phase adsorption of surface-active substances onto airborne nuclei is unknown. Certainly, laboratory studies to determine this possibility must be pursued.

ACKNOWLEDGMENT

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13. ABSTRACT

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Weather						
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